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CITATION:

Nakajima, T ...[et al]. Simultaneous production of spin-polarized ions/electrons based on two-photon ionization of laser-ablated metallic atoms. APPLIED PHYSICS LETTERS 2003, 83(11): 2103-2105

ISSUE DATE:

2003-09-15

URL:

http://hdl.handle.net/2433/50407

APPLIED PHYSICS LETTERS VOLUME 83, NUMBER 11 15 SEPTEMBER 2003

# Simultaneous production of spin-polarized ions/electrons based on two-photon ionization of laser-ablated metallic atoms

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(Received 28 April 2003; accepted 25 July 2003)

We demonstrate the simultaneous production of spin-polarized ions/electrons using two-color, two-photon ionization of laser-ablated metallic atoms. Specifically, we have applied the developed technique to laser-ablated Sr atoms, and found that the electron-spin polarization of $Sr^+$ ions, and accordingly, the spin polarization of photoelectrons is $64\% \pm 9\%$, which is in good agreement with the theoretical prediction we have recently reported [T. Nakajima and N. Yonekura, J. Chem. Phys. **117**, 2112 (2002)]. Our experimental results open up a simple way toward the construction of a spin-polarized dual ion/electron source. © *2003 American Institute of Physics.*
[DOI: 10.1063/1.1610800]

It is well known that photoelectrons coming out of a GaAs crystal irradiated by circular radiation is polarized. This is a standard method nowadays to produce polarized electrons, and used in a number of subfields such as surface physics, atomic and molecular physics, high-energy physics, etc. The drawback of this method is that the crystal can be seriously damaged by strong radiation. In a recent letter, multiphoton ionization of rare gas atoms has been proposed[1] for the production of polarized electrons, which is naturally damage free against laser intensity. Although the required wavelength is in the vacuum ultraviolet region, this is not a serious limitation nowadays because of the advent of the bright short-wavelength radiation source represented by free-electron lasers and high-order harmonics.

If the outgoing photoelectron is polarized, there is a possibility that the residual ions are electron-spin polarized as well, although how much the polarization can be is not *a priori* obvious. In order to clarify this problem, a theoretical study has been undertaken for alkaline-earth atoms, in which about 60%–90% electron-spin polarization of $Sr^+$ has been predicted upon two- and three-photon ionization of Sr.[2] Other interesting findings in Ref. 2 are that if all of the produced photoions are in the *s* state the spin polarization of photoions and photoelectrons are necessarily the same.

Not to mention the usefulness of spin-polarized electrons in various fields,[3] spin-polarized atoms/ions can be a powerful tool to study various dynamics at the surface.[4–8] Furthermore, electron-spin polarized ions with thermal energy would be very useful for the basic study of spin-dependent collisions in a gas phase. Alkaline-earth atoms may be most appropriate for this purpose, since, due to the single valence electron of the ion, the analysis of the dynamics can be greatly simplified under a number of situations.

Thus, the use of alkaline-earth atoms rather than a GaAs crystal or rare gas as a target can be an interesting alternative, since both spin-polarized ions and electrons will be simultaneously available upon photoionization. Although the fact that alkaline-earth atoms have rather high melting points may seem to result in a low number density of atoms, the difficulty can be easily overcome by the use of the laser-ablation technique, which has been employed as a simple and compact atom/molecular source for the various spectroscopic studies.[9,10]

In this letter, we demonstrate the simultaneous production of spin-polarized ions/electrons based on two-color, two-photon ionization of laser-ablated Sr atoms, which will serve as an important cornerstone toward the construction of a spin-polarized dual ion/electron source. We would like to emphasize that, although optical pumping or spin exchange is the standard technique to produce electron-spin polarized ions,[8,11–13] we employ neither of them. As explained in Ref. 2, in order for the alkaline-earth atom to emit a spin-polarized photoelectron, it is essential that the ionization takes place from a triplet state. This is because radiation interacts only with the orbital angular momentum of the electron(s), and we need spin-orbit interactions to transfer the polarization of orbital angular momentum to that of spin angular momentum. On the contrary, there is no spin-orbit interaction in a singlet state.

Figure 1 shows the experimental setup. A solid Sr disk with natural isotope abundance is placed in a vacuum chamber whose background pressure is maintained to be $\sim 1 \times 10^{-4}$ Pa by two turbomolecular pumps installed in series. A fundamental laser beam of a Nd:YAG laser (Newwave Research, Tempest, 1.06 $\mu$m, 10 ns pulse duration, 10 Hz) is focused with a 25 cm focal length lens onto the Sr disk with normal incidence. The power density of the ablation laser onto the disk is approximately 2 J/cm$^2$. From the spectro-

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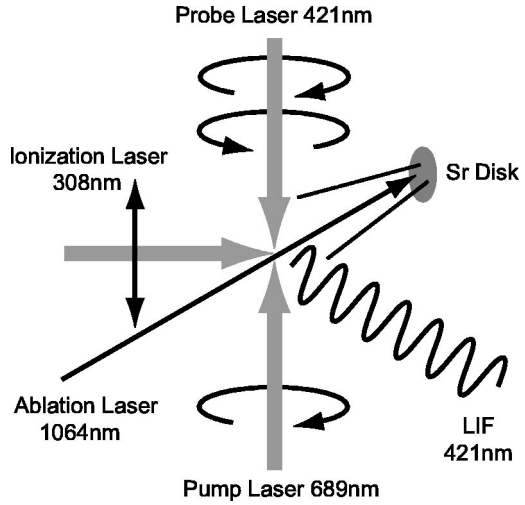


FIG. 1. Experimental setup.

scopic analysis using a monochromator and a photomultiplier tube, we have confirmed that the ablation plume ceases in a few $\mu$s. At 50 $\mu$s after the ablation pulse, the output of a pulsed dye laser (Lambda Physik FL3002, 18 ns pulse duration) pumped by a XeCl excimer laser (Lambda Physik MSC103, 18 ns pulse duration) and the output of the excimer laser itself are introduced into the interaction region, which is 2 cm away from the disk surface. The wavelength of the dye laser is 689 nm so that one-photon resonant, two-photon ionization takes place via $5s5p\ ^3P_1$ (see Fig. 2) by absorbing each of the 689 and 308 nm photons. The dye laser and the excimer laser have right-circular and linear polarization, respectively, through the quarter-wave plate and the polarizing beamsplitter cube. As shown in Fig. 1, they are in the cross-beam geometry at right angle so that the transitions illustrated in Fig. 2 take place. We estimate that the intensities of the pump and ionization lasers are 44 kW/cm$^2$ and 2.5 MW/cm$^2$, respectively, resulting in the photoionization of $\sim$25% of atoms in the interaction region.

Thus produced $Sr^+$ ions are in the ground $5s\ ^2S_{1/2}$ state. By introducing the third laser (Lambda Physik FL3002) at 421 nm, which corresponds to the $Sr^+\ 5s\ ^2S_{1/2}-5p\ ^2P_{1/2}$ transition and counterpropagates to the pump laser beam, ions can be detected from the laser-induced fluorescence (LIF) at the same wavelength. The LIF is collected with a

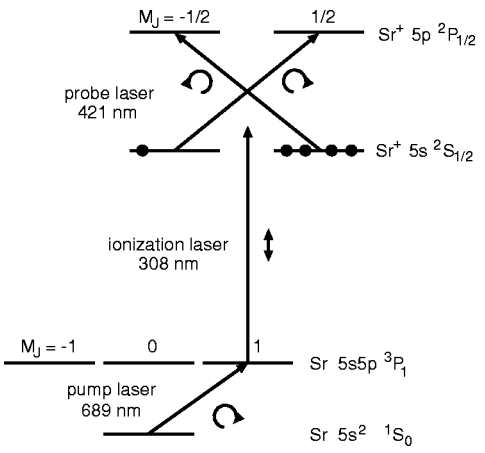


FIG. 2. Level scheme.

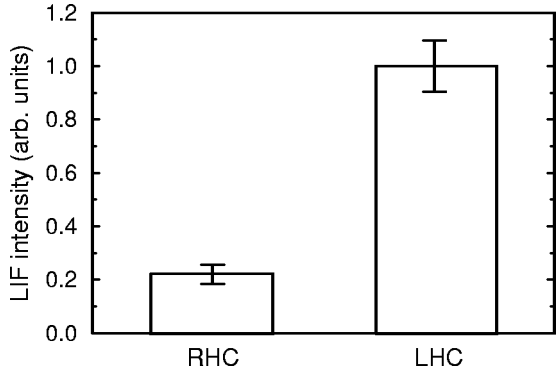


FIG. 3. LIF intensity probed by the right-/left-circularly polarized probe laser, corresponding to the $Sr^+\ 5p\ ^2P_{1/2}-5s\ ^2S_{1/2}$ transition.

pair of 10 cm focusing lenses and imaged onto the photomultiplier tube. The time-resolved LIF signal is then amplified by a fast preamplifier and sent to a digital oscilloscope. Averaging is performed over 200 shots through the box-car integrator.

In order to ensure that the ions in the interaction region are indeed produced by absorbing each of the 689 and 308 nm photons, we turn off either one of the pump and ionization lasers, and then both of them, to find that the LIF signal becomes a background (but nonzero) level in all cases. This undesired background signal is attributed to the residual ions produced by laser ablation itself. Due to the recombination processes, etc., the number of residual ions decreases at longer pulse delay after the ablation at the expense of a smaller number of neutral atoms. As a matter of fact, this is the reason why we have chosen the delay time of 50 $\mu$s after the ablation pulse, so that the contribution of the background LIF signal due to the residual ions becomes less than 5% of the LIF signal. For further reduction of the residual ions, we have installed a pair of electrodes between the disk and the interaction region, and applied a pulsed electric field of 150 V/cm to deflect residual ions. The pulsed electric field is completely turned off at 20 $\mu$s after the ablation, and we have confirmed that the background signal has been eliminated to $<1$% of the LIF signal.

The degree of electron-spin polarization of $Sr^+\ 5s\ ^2S_{1/2}$ ions is experimentally determined from the ratio of the LIF intensities by the right-/left-circularly polarized probe laser, i.e.,

$$P=\frac{I_{\mathrm{LHC}}-I_{\mathrm{RHC}}}{I_{\mathrm{LHC}}+I_{\mathrm{RHC}}}, \tag{1}$$

where $I_{\mathrm{RHC}}$ and $I_{\mathrm{LHC}}$ are the LIF intensities by the right-/left-circularly polarized probe laser. The sense of circular polarization for the probe laser is adjusted by rotating the quarter-wave plate. Because of the geometry of the laser beams and the laser polarization we have chosen, spatial distribution of the LIF emission is uniform in the horizontal plane. Clearly, no calibration for the detection efficiency of $I_{\mathrm{RHC}}$ and $I_{\mathrm{LHC}}$ is needed.

In Fig. 3, we show the LIF intensity probed by the right-/left-circularly polarized probe laser, corresponding to the $Sr^+\ 5p\ ^2P_{1/2}-5s\ ^2S_{1/2}$ transition. In reality, 7% of Sr atoms (and, therefore, ions) with natural isotope abundance have nuclear spin of $I=9/2$, and therefore, we have to take into

account the hyperfine depolarization[14] to estimate the degree of spin polarization. For our specific case, however, the effect of the hyperfine depolarization may be neglected for the zeroth-order approximation, since the hyperfine coupling time of the $Sr^+$ $5p\ ^2P_{1/2}$, is comparable to its spontaneous decay time and the hyperfine splitting of the transition is smaller than the probe laser bandwidth. After these considerations, the degree of spin-polarization is found to be 64% $\pm$9%, which agrees well with the theoretical prediction.[2] From Eqs. (16), (B11), and (B12) in Ref. 2, the theoretical spin-polarization $P_{\mathrm{th}}$ for the scheme considered in this letter is written as

$$P_{\mathrm{th}}=\frac{0.1665|R_{5p}^{ks}|^2+0.1332|R_{5p}^{kd}|^2}{0.1665|R_{5p}^{ks}|^2+0.2336|R_{5p}^{kd}|^2}, \tag{2}$$

in which $R_{5p}^{ks}$ and $R_{5p}^{kd}$ are the radial matrix elements from $5s5p\ ^3P_1$ to the $5sks$ and $5skd$ continua where $k$ stands for the wave vector of photoelectrons. Needless to say, the values of $R_{5p}^{ks}$ and $R_{5p}^{kd}$, and naturally the ratio between them, vary as a function of photoelectron energy. From the above expression, we find that the maximum and the minimum possible values of $P_{\mathrm{th}}$ are $0.57<P_{\mathrm{th}}<1$. The small difference between the experimental and the theoretical values of electron-spin polarization indicates that the ratio of $R_{5p}^{ks}$ and $R_{5p}^{kd}$ employed for the theoretical calculation is slightly off. It should be clear now that, if a different wavelength is used for the ionization laser, the degree of spin polarization can take a different value within $0.57<P<1$. In particular, it is known that the spin polarization of photoelectrons rapidly changes in the vicinity of autoionization resonance.[1,15] Therefore, we may expect that the ratio of the $s$ and $d$ wave in the continuum could rapidly change as well near an autoionization resonance. It would be interesting to study the variation of spin polarization near the autoionization resonance with the help of the optical detection technique developed in this work.

In conclusion, we have demonstrated the simultaneous production of spin-polarized ions/electrons based on two-color, two-photon ionization of laser-ablated Sr atoms. The degree of electron-spin polarization of $Sr^+$ ions has been found to be 64% $\pm$9% from the polarization analysis of the laser-induced fluorescence, which agrees well with the theory. In particular, if all the produced photoions are in the $s$ state which is indeed our case, there is an exact one-to-one correspondence between the spin polarization of photoions and photoelectrons, as found in a recent theoretical paper.[2] This means that the spin polarization of photoelectrons is necessarily 64% $\pm$9% as well without any ambiguity, and the degree of spin polarization of photoelectrons can be easily monitored, without the use of a Mott detector, through the laser-induced fluorescence of photoions. Finally, the technique developed in this letter opens up a way toward the construction of a spin-polarized dual ion/electron source.

This work was supported by the Grant-in-Aid for scientific research from the Ministry of Education and Science of Japan.

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